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Exciton localization by clusters in diluted bulk InGaN and two-dimensional ZnCdSe solid solutions

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Abstract. The photoluminescence (PL) bands of MOCVD grown double heterostructures (DHS) GaN/InGaN/GaN with In content in the range 0.4–2% as well as MEE grown ZnSe/CdSe/ZnSe QWs with 0.3–1.5 ML CdSe nominal thicknesses show a fine structure which can be attributed to small In or Cd clusters, respectively, occurring in cation sublattices at random filling of the lattice sites. This fine structure is smoothed away at further concentration increase in both systems. At the lowest concentration the exciton motion in these systems has a character of 3D and 2D percolation over the "impurity" bands formed by clusters of size $s = 2$ in both cases. We have found that similarity of InGaN and ZnCdSe PL-band shapes despite the different dimensionality of objects (3D DHS and 2D QWs) is due to a different perturbation strength produced by In and Cd, respectively.

Introduction

Solid solution grown from constituents with highly mismatched lattices can be considered as "ideal" or random alloy, at best, at low concentration only. On the other hand, the conventional treatment of disorder effect on the electron states in a solid solution assumes usually its random character. In the low concentration limit the random distribution of the diluted atoms can be described by clusters of relatively small size [ ]. As a result, the localization of exciton occurs due to perturbation of the potential relief by randomly distributed clusters. The shape of the density of states (DOS) of localized states is very sensitive to the size and number of clusters which are responsible for the localization. For 3D systems the low concentration limit is most interesting in cases when the perturbation strength of diluted atoms is large enough for exciton localization by the clusters of small size. In these cases the cluster structure of DOS can be directly observed in the luminescence spectra [ ].

2D exciton states which can be realized in QWs formed by solid solution are much more sensitive to the disorder and, therefore, a relatively weak perturbation can be sufficient for the cluster localization of exciton. Here two important problems are encountered: (i) what is the minimum concentration of the diluted atoms necessary to form a QW, and (ii) how to distinguish the mobile and localized excitons in the case when the fluctuations can not be considered as a small correction.

We have studied PL and excitation of PL (PLE) spectra of two systems: (i) MOCVD grown GaN/InGaN/GaN double heterostructures (DHs) with a well width of 100 nm and an In content from 0.4% to 2%, and (ii) single QWs formed by a MEE cycled deposition of CdSe (by 0.3 ML per cycle) in ZnSe matrices with the number of cycles 1, 2 and 5

(for more growth details see [3, 4], respectively). The 441.6 nm or 325 nm He-Cd laser lines were used to excite PL spectra of ZnCdSe and InGaN systems, respectively. PLE spectra were excited by a second monochromator with Xe-lamp. For both systems we have found that in the composition range under investigation the PL spectra posses a fine structure, which can be attributed to exciton localization by clusters of 2 or 3 atoms of the narrow gap component. The number of such clusters obeys the random distribution statistics. For In content exceeding ≈ 0.02 in InGaN solid solutions as well as for Cd content in ZnCdSe more than $5 \cdot 0.3$ ML this fine structure in the PL spectra is smoothed out and the Stokes shifts between PL and PLE spectra becomes larger than that expected for the random statistics [5].

1. Cluster structure of InGaN luminescence band in the limit of low In concentration

The luminescence spectra of InGaN epilayers in Figs. 1(a,b) show a structure which becomes less pronounced with the In content increase and disappears completely already at 2% of In (Fig. 1(c)). Due to this structure the PL in Figs. 1(a,b) differs considerably from the usual structureless spectra of concentrated solid solutions with the Urbach tails of localized states.

1. To account for the observed structure of PL bands we have assumed a random distribution of In atoms over the cation sublattice. Neglecting the difference between the sublattice geometry of hexagonal InGaN and that of a fcc lattice we have used the percolation theory for fcc lattice to find the number of clusters of different size. Using the approach described in [1] we have calculated the exciton density of states (DOS) (curves 1), spectral density of states (SDOS) (curves 2), and SDOS of radiative states (curves 3 in Fig. 1). The cluster structure can appear at low concentrations if the perturbation strength produced by substituting atoms is large enough and the localized states split off for the clusters which size does not exceed $s = 3$. The number of space configurations of clusters of larger size increases rapidly [6] and a dependence of the localization energy on the cluster configuration suppresses appearance of resolved luminescence structure for such clusters. The model fit of experimental data presented in Fig. 1 shows that the structure of PL spectra of InGaN solid solution can be assigned to clusters with $s = 2, 3$.

2. The following characteristics of an InGaN solid solution were obtained as a result of calculations.

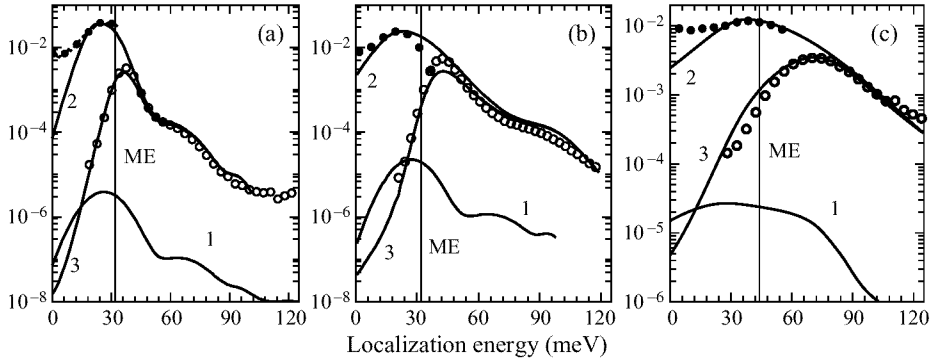

Fig. 1. PL (open circles) and PLE (solid circles) spectra of GaN/InGaN/GaN DHS with In content 0.4% (a), 1% (b) and 2% (c) (symbols). Solid lines are the calculated DOS (1), spectral DOS (SDOS) (2), and SDOS of radiative states (3). Vertical lines ME mark the mobility edge positions.

a) The potential well depth created for exciton by single In atom in GaN lattice is about $\Delta \approx 400$ meV.
b) The ratio of the potential amplitude Δ to the critical perturbation which splitts off the localized state with zeroth localization energy is close to unity ($\Delta/E_{cr} \approx 0.9$) which means the strong scattering regime already for a single In atoms.
c) The bowing parameter for composition dependence of the band gap is large and equal to ≈ 3 eV.
d) For the studied compositions the percolation threshold (ME) is situated in the energy range where the clusters of two In atoms are responsible for the DOS and moves toward the deeper localization energies with the increase in In concentration.

2. Cluster structure of PL spectra of QWs formed by ZnCdSe solid solution at small Cd concentrations

1. The PL and PLE spectra of QWs with nominal Cd thickness of 1, 2, and 5 cycles by 0.3 ML are shown in Figs. 2(a,b,c), respectively. For these samples we estimated the average Cd content in broadened QWs as 2.5, 5 and 12.5% which is in good agreement with the data of [7]. The PL spectra of the first two samples show a structure which can be attributed to the clusters with $s = 2$ and 3. This structure disappears with the Cd content increase and at 12.5% Cd it is not observable. The calculations performed have allowed us to estimate also the potential amplitude of Cd in the ZnSe matrix as $\Delta \approx 150$ meV and the ratio $\Delta/E_{cr} \approx 0.3$ (E_{cr} is a characteristics of the 3D ZnCdSe solid solution). The obtained small values of Δ/E_{cr} and Δ explain why the 3D Urbach tail is not detected experimentally in this solid solution [8]. Hence, the observed properties of PL spectra of QWs are due entirely to the 2D character of exciton states in this system.

2. Taking into account that the QW width amounts about 10 ML [7], we have used in calculations of the number of clusters also the 3D statistics for the fcc lattice. The results can be formulated as follows:
(a) The fine structure of PL band is concerned with the clusters of size $s = 2$ and 3.
(b) The mobility edge position occurs in the energy range of the $s = 2, 3$ DOS maxima. Therefore, as in the previous InGaN case the exciton motion has a character of percolation over an "impurity band" formed by these clusters.
(c) The position of the mobility edge in the QW formed by 0.3 ML of Cd is close to barrier exciton SDOS maximum while the SDOS of the 2D excitons corresponds to the localized

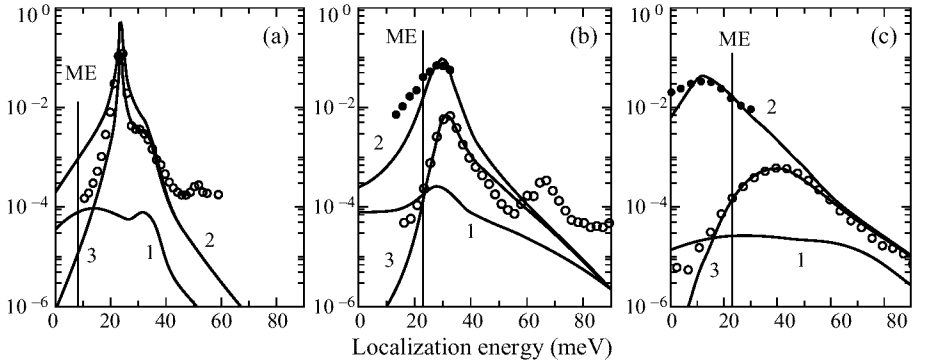

Fig. 2. Similar to that in Fig. 1 but for ZnCdSe QWs with nominal CdSe thicknesses 0.3 ML (a), 2 · 0.3 ML (b) and 5 · 0.3 ML (c).

exciton states. So we can conclude that the concentration of Cd in 0.3 ML QW is close to the critical one at which the QW is just formed.

Summary

We have found a fine structure of PL spectra of diluted bulk InGaN and 2D ZnCdSe solid solutions and have attributed it to the DOS structure caused by statistical clusters consisting of 2 or 3 atoms of narrow gap component. The perturbation potentials of In in GaN and Cd in ZnSe are estimated. It is shown that a single In atom in GaN produces a perturbation which is almost sufficient for the splitting off the localized state in 3D case, while the perturbation of Cd atom in ZnSe matrix gives a localized state only in the systems of reduced dimensionalities.

Acknowledgments

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